

MODELING CHEMICAL SPECIATION AND RELEASE FROM CEMENT STABILIZED WASTES USING LEACHXS

Hans van der Sloot, Hans Meeussen, Andre van Zomeren

ECN, Environmental Impact Assessment Group, The Netherlands

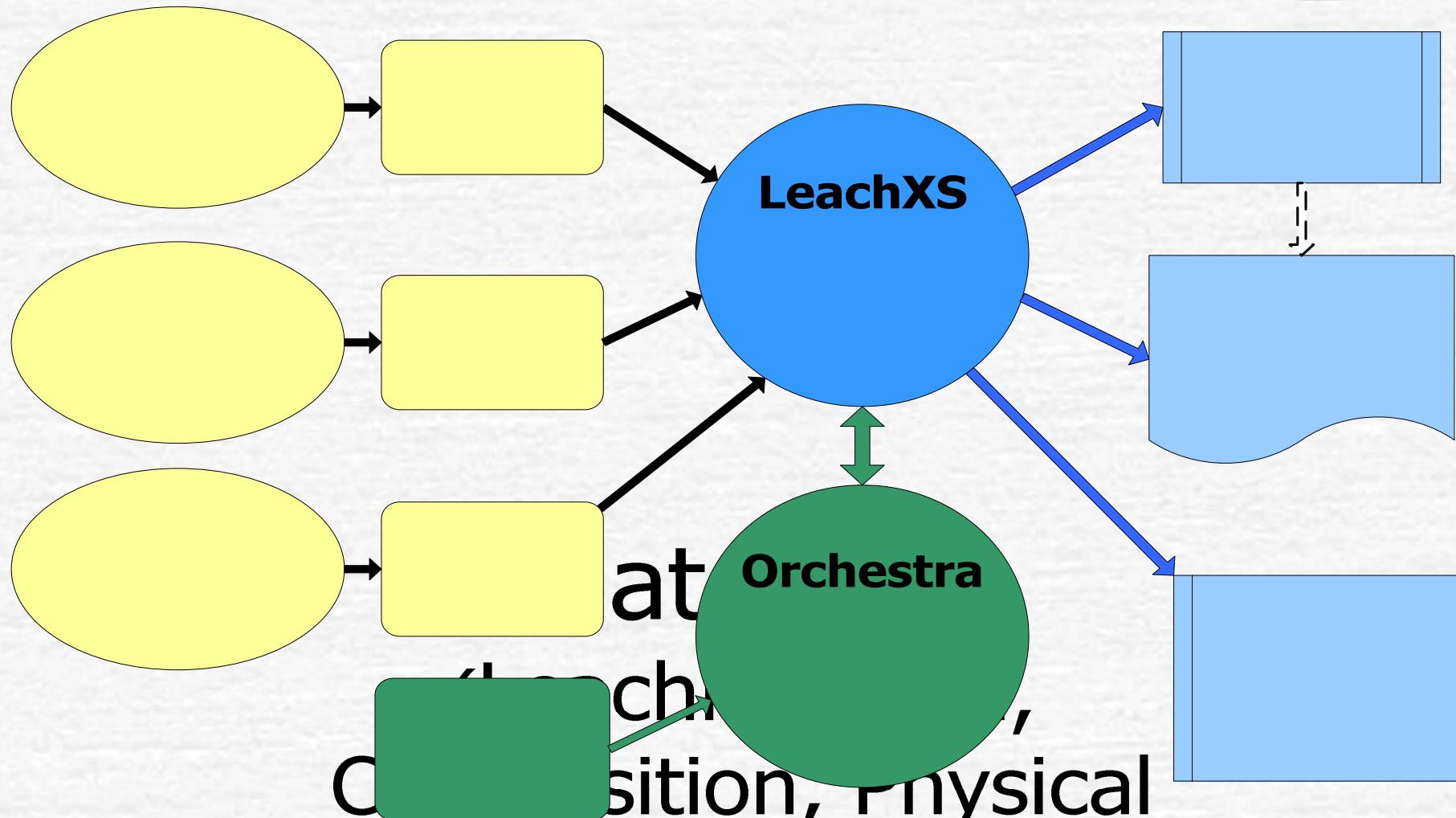
**CEMENTITIOUS MATERIALS for WASTE TREATMENT, DISPOSAL,
REMEDIATION and DECOMMISSIONING WORKSHOP
TUESDAY - December 12, 2006**



OUTLINE

- ☞ Leachxs – Orchestra introduction
- ☞ Scenario description stabilised waste
- ☞ Testing
- ☞ Modelling test results
- ☞ Impact modelling
- ☞ External influences e.g. carbonation oxidation
- ☞ Conclusions

LeachXS Structure

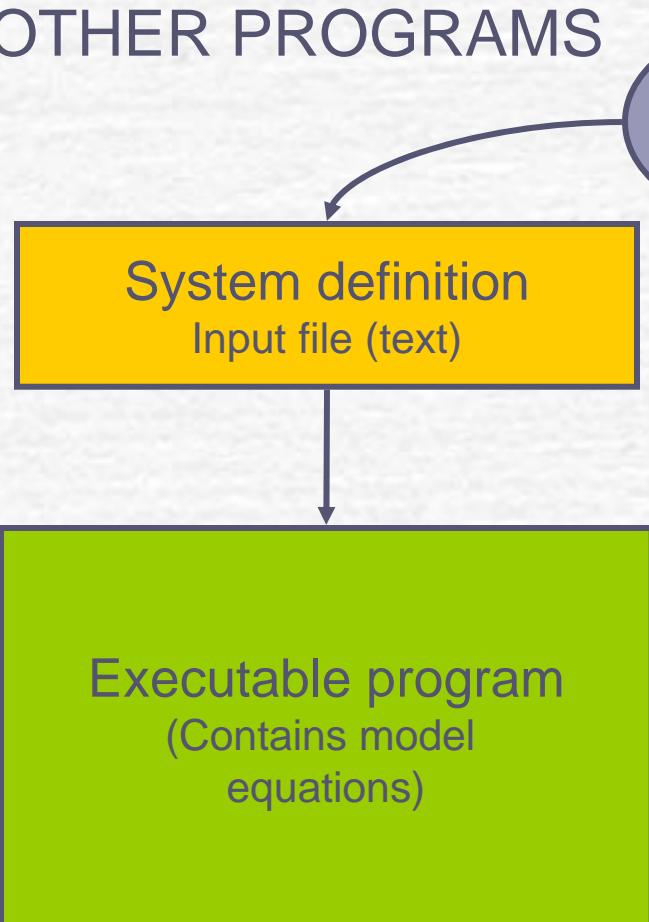


at
ch
c
characteristics)

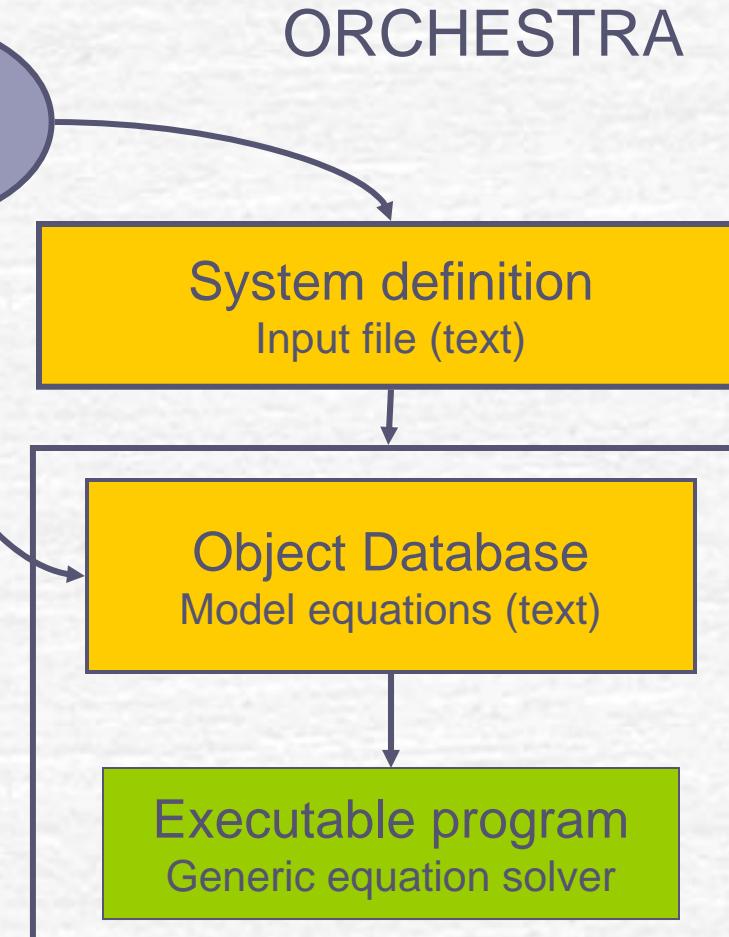
Jointly developed by ECN, Vanderbilt University and DHI

ORCHESTRA Features

OTHER PROGRAMS



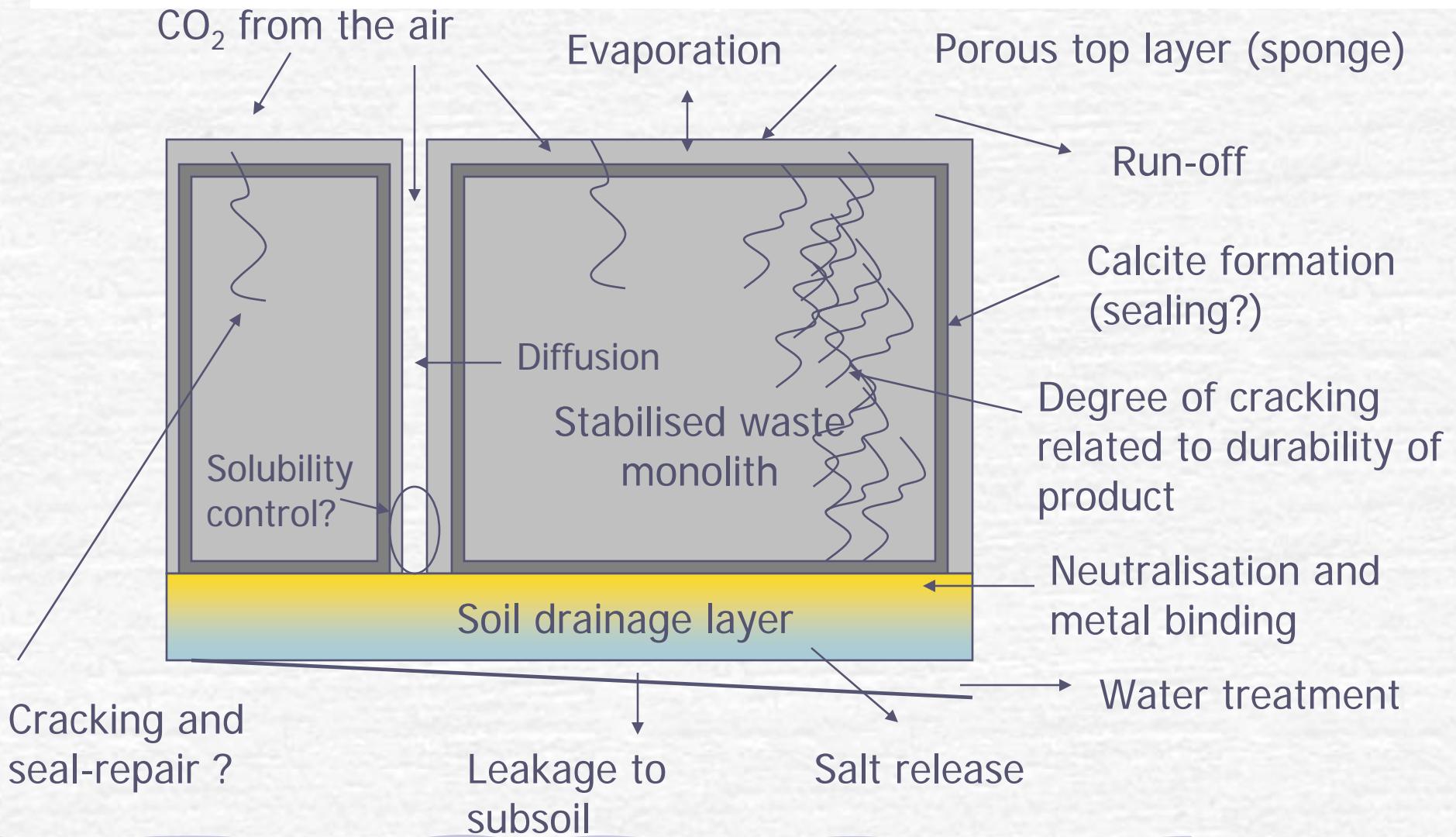
ORCHESTRA



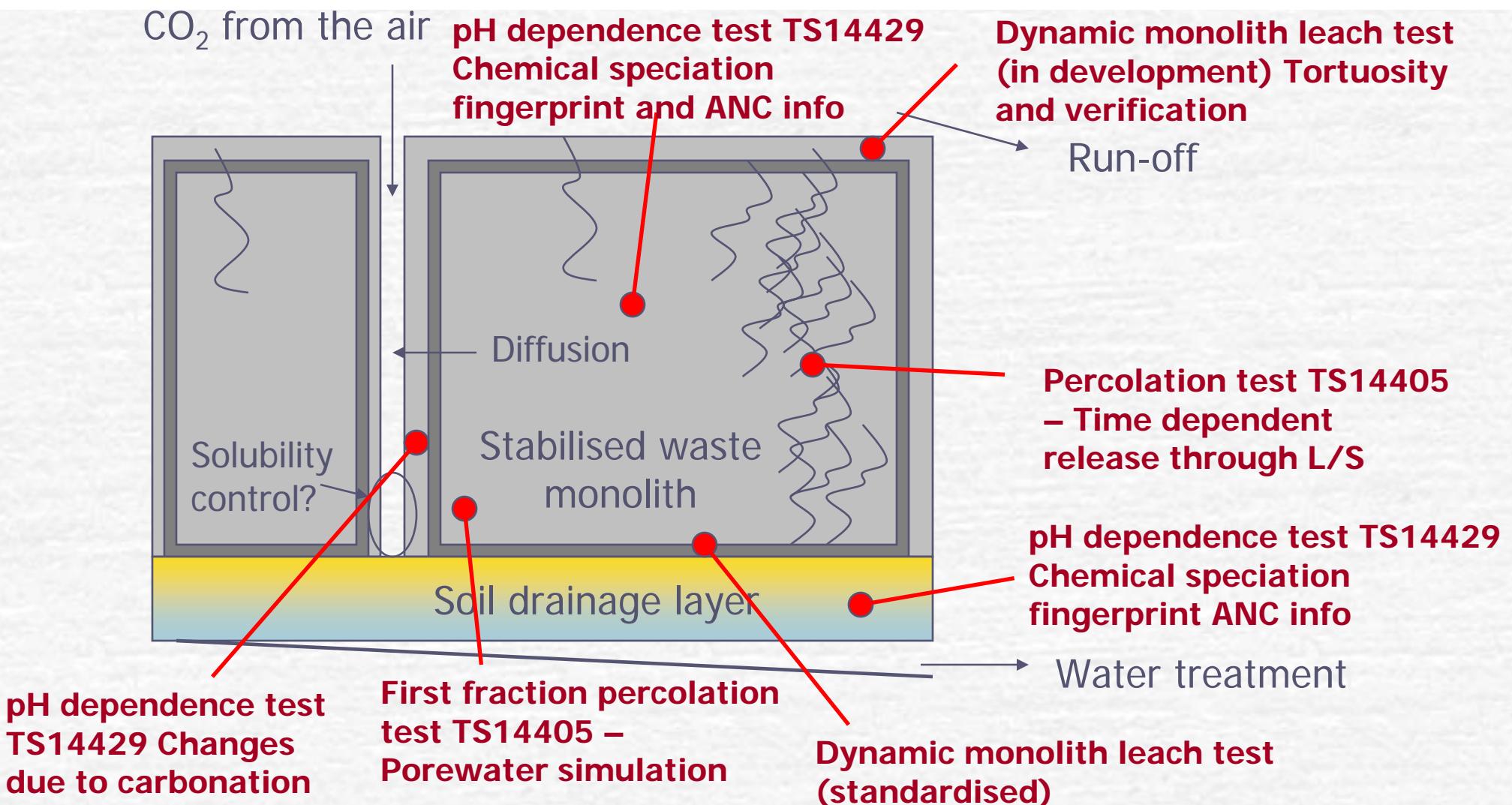
Predefined Orchestra-LeachXS models

- ✓ Using multi-component chemical interaction model
- ✓ Including:
 - 45 Elements / Master species
 - Literature aqueous chemical complexation reactions (NIST/MINTEQV4)
 - Literature adsorption models (Fe Al oxides : Dzombak & Morel 1990; Organic Matter Nica – Donnan: Kinniburgh et al 1996, Clay Ion exchange: Donnan model)
 - Solid solution (ideal) for Ettringite + oxyanions
 - Activity models: Davies equation and modified Pitzer (Samson and Lemaire 1999)
- ✓ Bulk (multi element) export functions to MS Excel (tables and graphs)
- ✓ Export function of graphs from MS Excel to MS Word

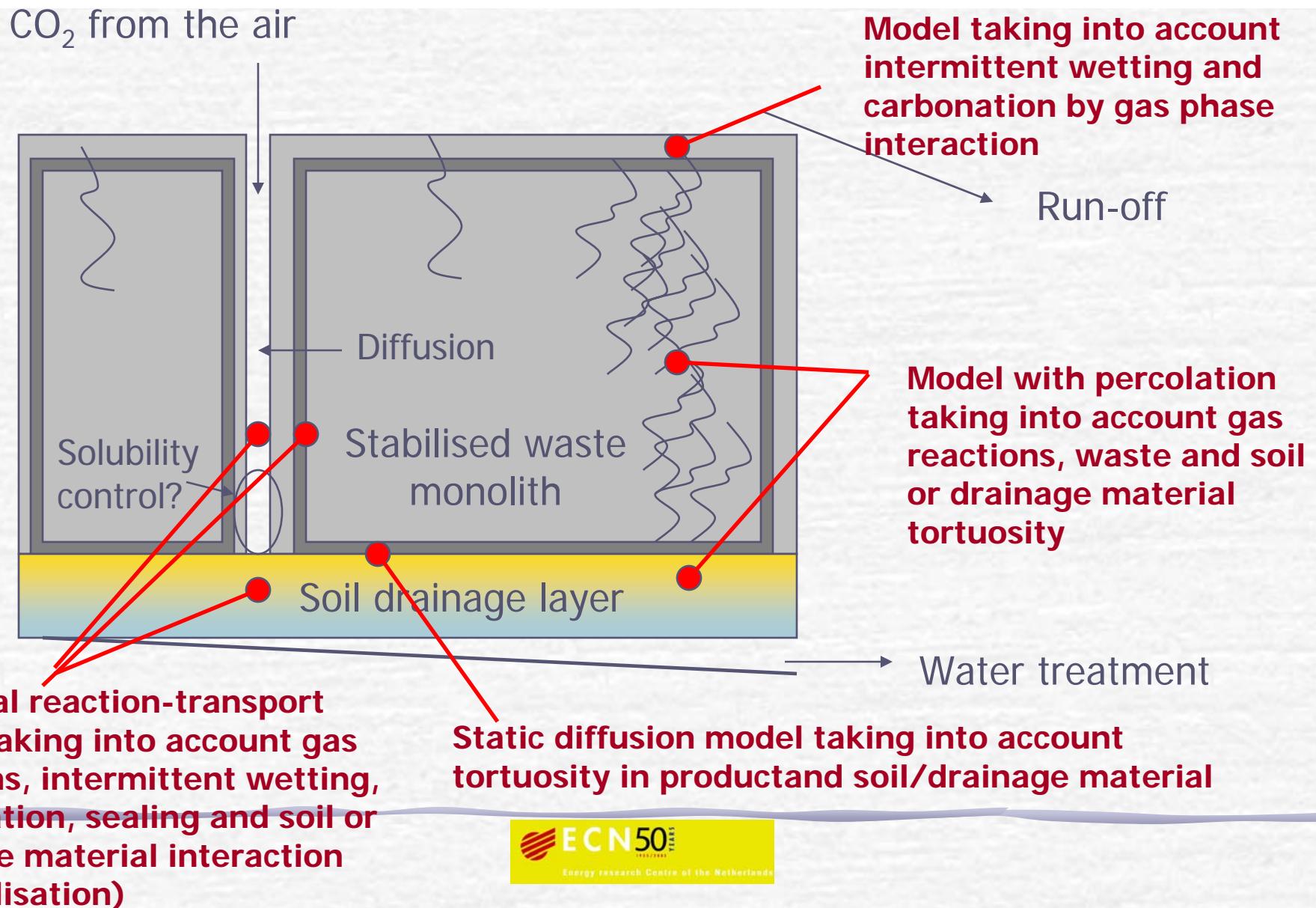
SCENARIO DESCRIPTION: IDENTIFICATION OF PROCESSES IN STABILISED WASTE DISPOSAL



SCENARIO DESCRIPTION: IDENTIFICATION OF TESTS TO ASSESS RELEASE FROM STABILISED WASTE

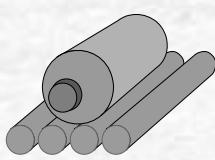


SCENARIO DESCRIPTION: IDENTIFICATION OF MODEL COMPONENTS TO DESCRIBE RELEASE

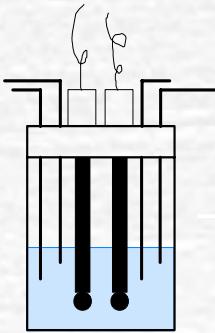


CHARACTERISATION LEACHING TESTS

GRANULAR MATERIALS



or



pH DEPENDENCE
TEST : BATCH
MODE ANC
TS 14429 or
COMPUTER
CONTROLLED
TS 14997



PERCOLATION
LEACHING TEST
(TS 14405)

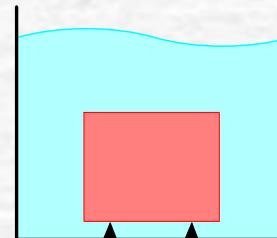
MONOLITHIC MATERIALS

Crushed

Same as granular

+

Intact



TANK LEACH
TEST
(MONOLITH)
and
COMPACTED
GRANULAR
LEACH TEST.

EN 12920

- Scenario Description
- Material characterizat ion
- Controlling factors
- Modelling leaching
- Validation verificat ion
- Evaluation
- Conclusions

Chemical speciation aspects

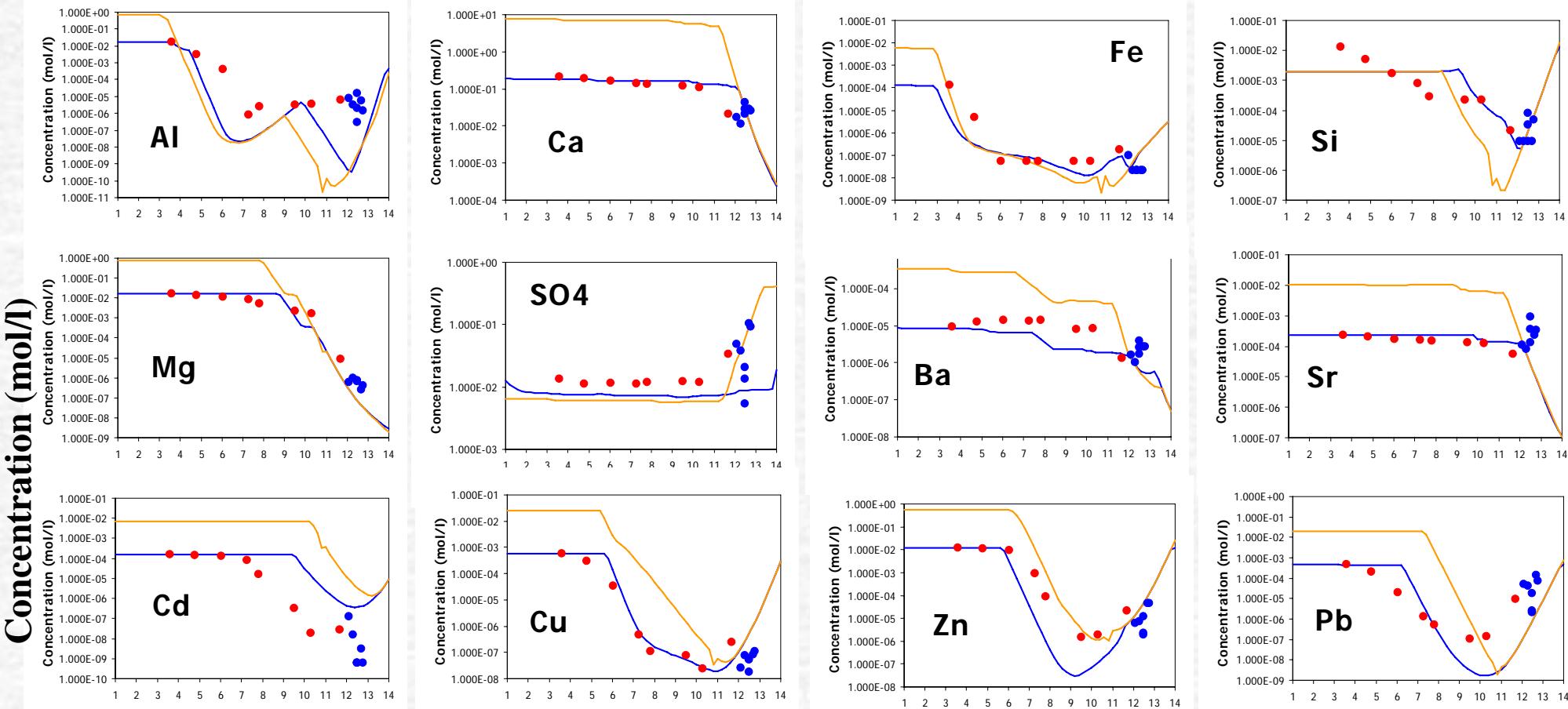
Time dependent release

CEN/TC292, ISO TC190, CEN/ TC345,
CEN/TC351, SW846

STEPS IN CHEMICAL SPECIATION MODELING

1. pH dependence leaching test on granular material or size reduced monolithic material for chemical speciation purposes
2. measurement of release from granular materials in a percolation test or from monolithic specimen according to a type of tank test
3. speciation modelling using LeachXS, a database-coupled version of the modelling environment ORCHESTRA, to identify relevant mineral phases (SI-indices)
4. prediction of leaching behaviour in a pH dependence test based on selected minerals and other relevant phases (Fe, Al, clay, DOC, etc) providing a chemical speciation fingerprint
5. this resulting speciation is used as input for the chemical reaction/transport modelling to describe the release from a percolation test of from a monolithic specimen (tank test simulation)
6. Full mechanistic model the field scenario with external factors (carbonation, oxidation, biologically mediated reactions) and realistic estimates of infiltration.

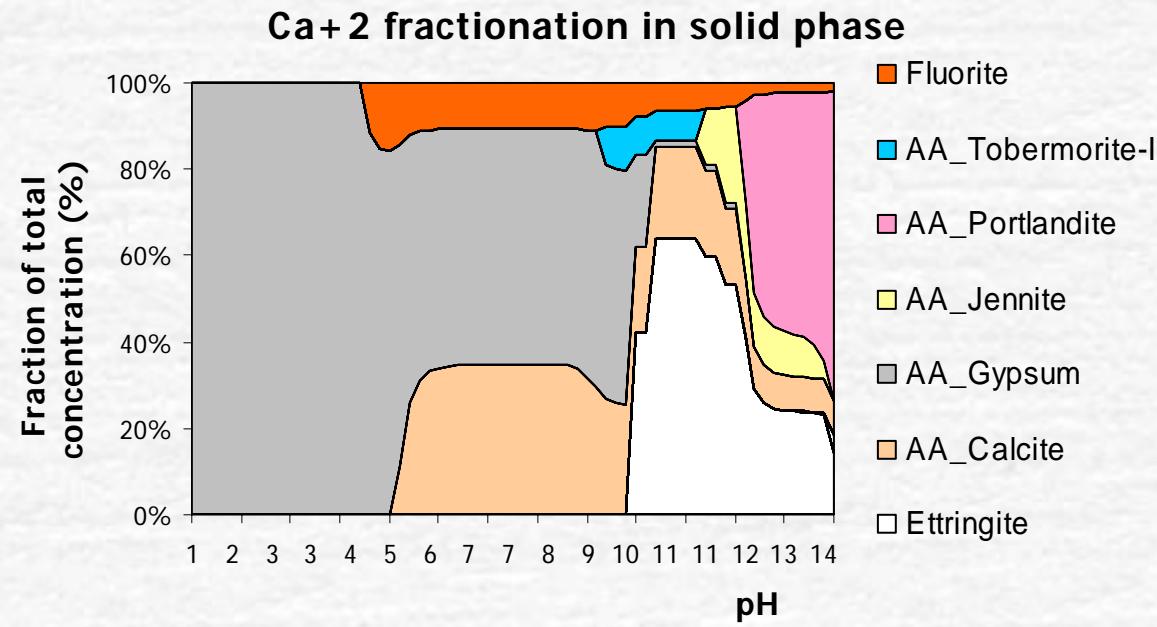
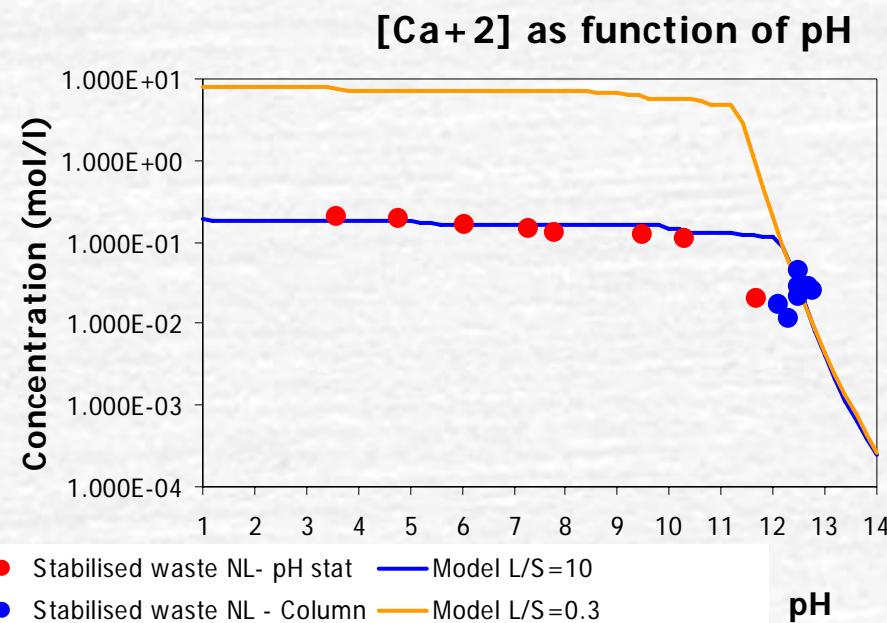
MULTIELEMENT PREDICTIVE MODELLING OF pH DEPENDENCE TEST DATA AT L/S=10 and L/S= 0.3



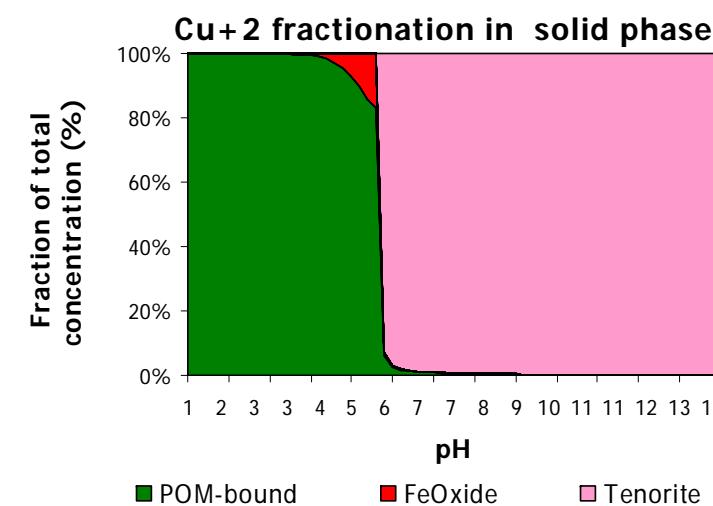
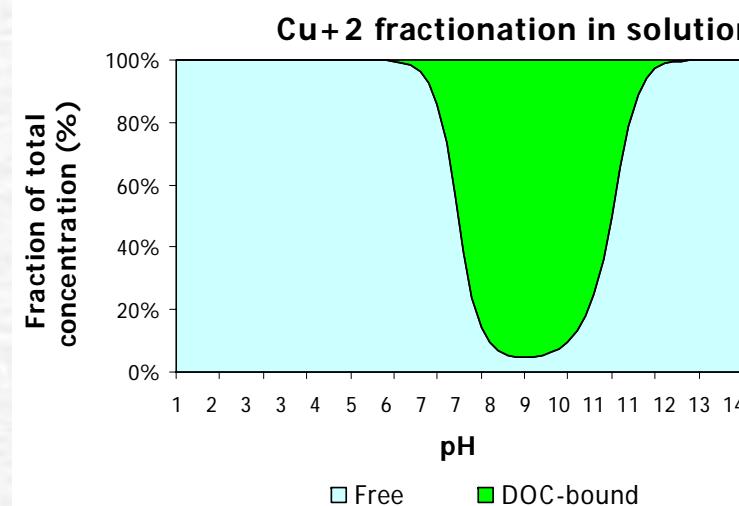
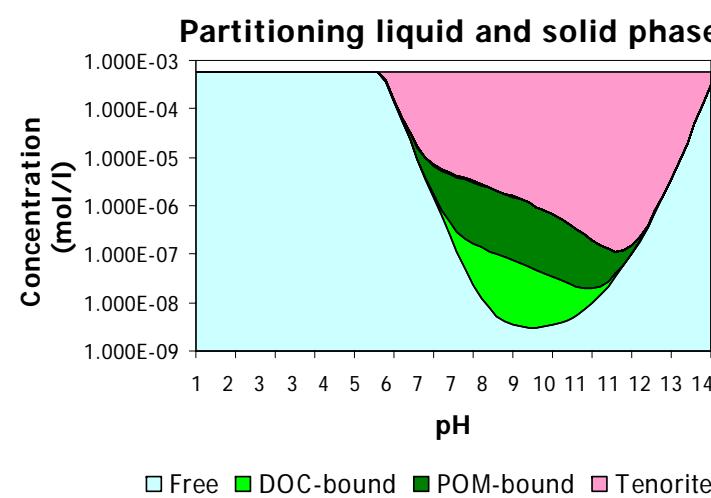
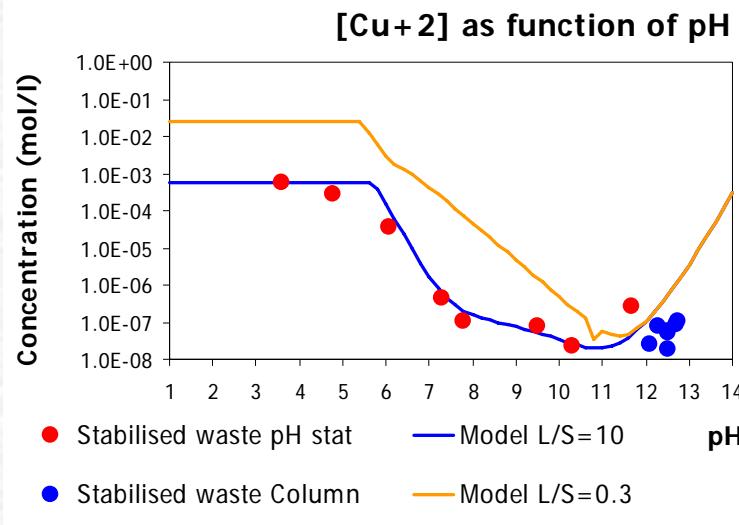
Red dots – pH stat L/S=10; Blue diamonds – Column test L/S=0.1-10;
Blue line – L/S=10 prediction; Yellow line – L/S=0.3 prediction

pH

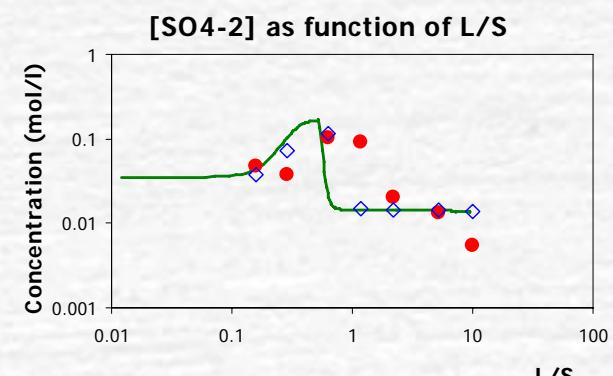
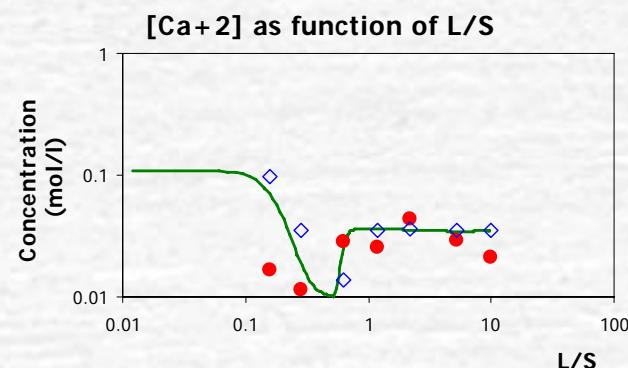
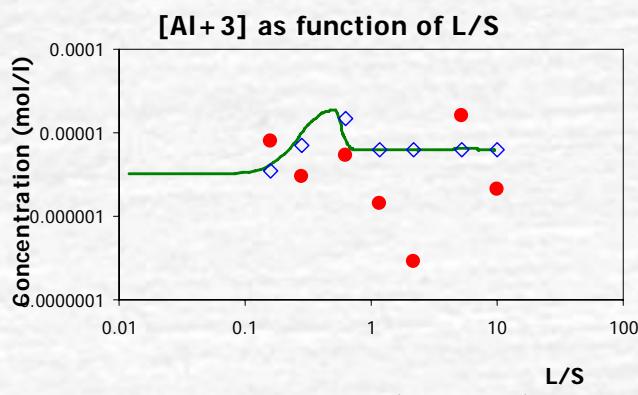
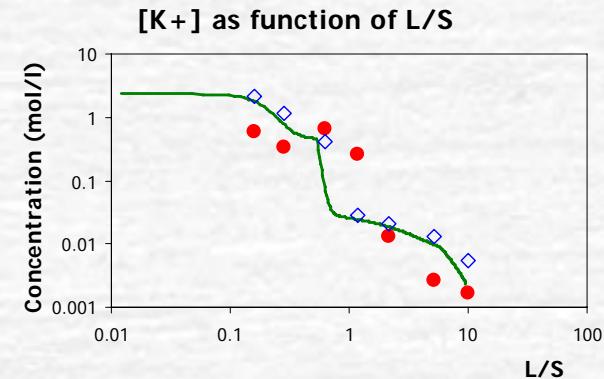
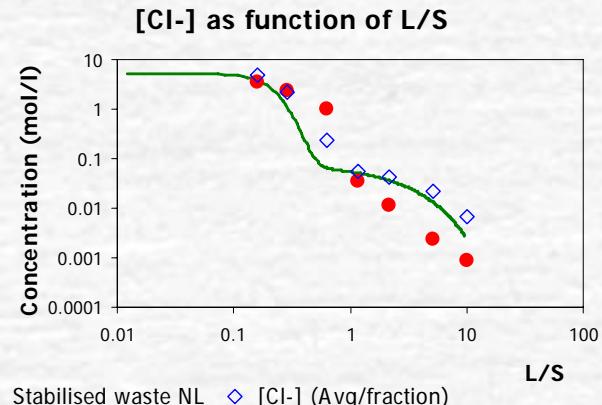
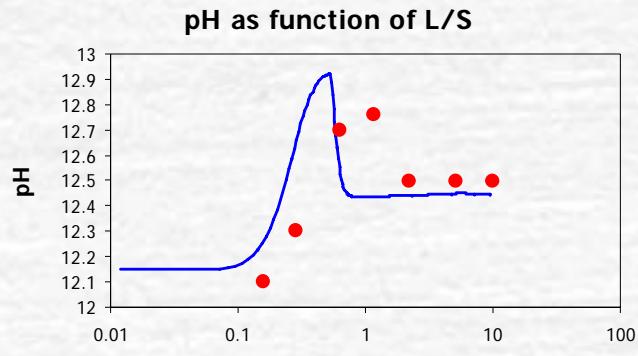
PARTITIONING FROM MULTIELEMENT PREDICTIVE MODELLING OF pH DEPENDENCE TEST DATA AT L/S=10 and L/S= 0.3



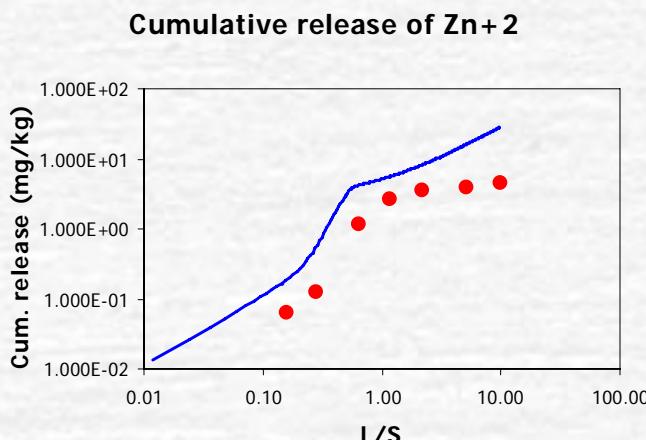
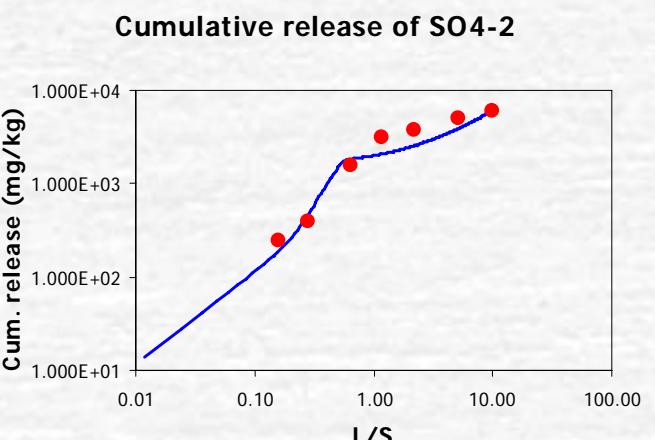
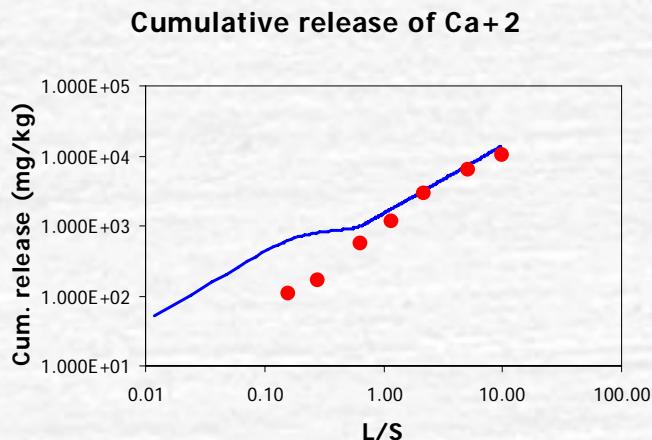
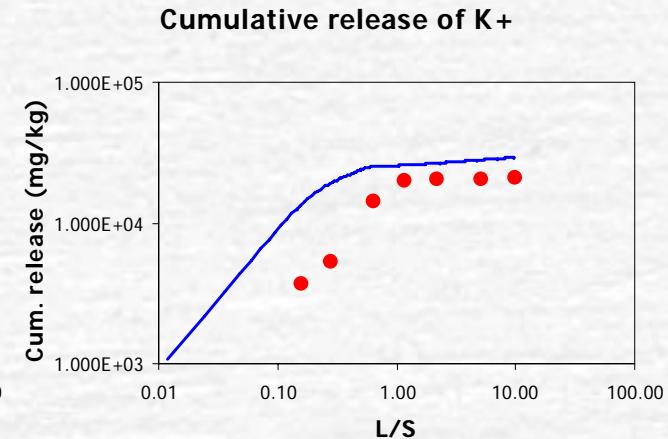
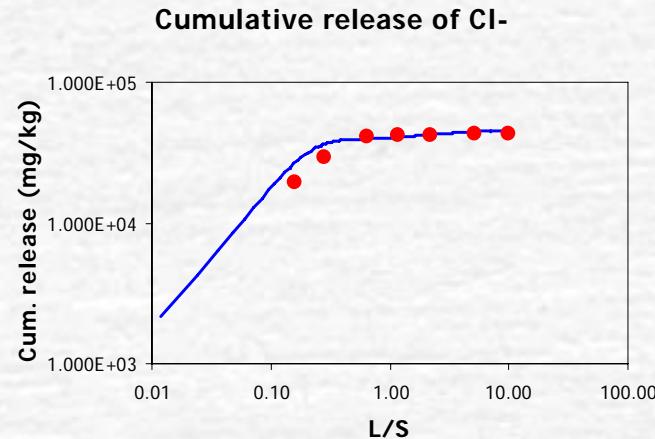
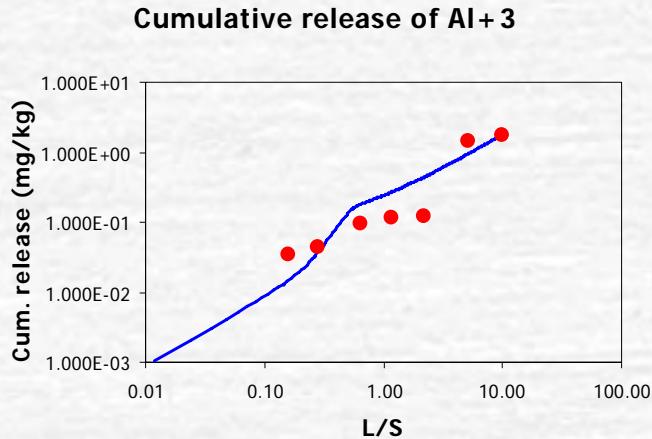
PARTITIONING FROM MULTIELEMENT PREDICTIVE MODELLING OF pH DEPENDENCE TEST DATA AT L/S=10 and L/S= 0.3



FULL MECHANISTIC MULTIELEMENT PREDICTIVE MODELLING OF PERCOLATION TEST DATA FOR CRUSHED STABILISED WASTE

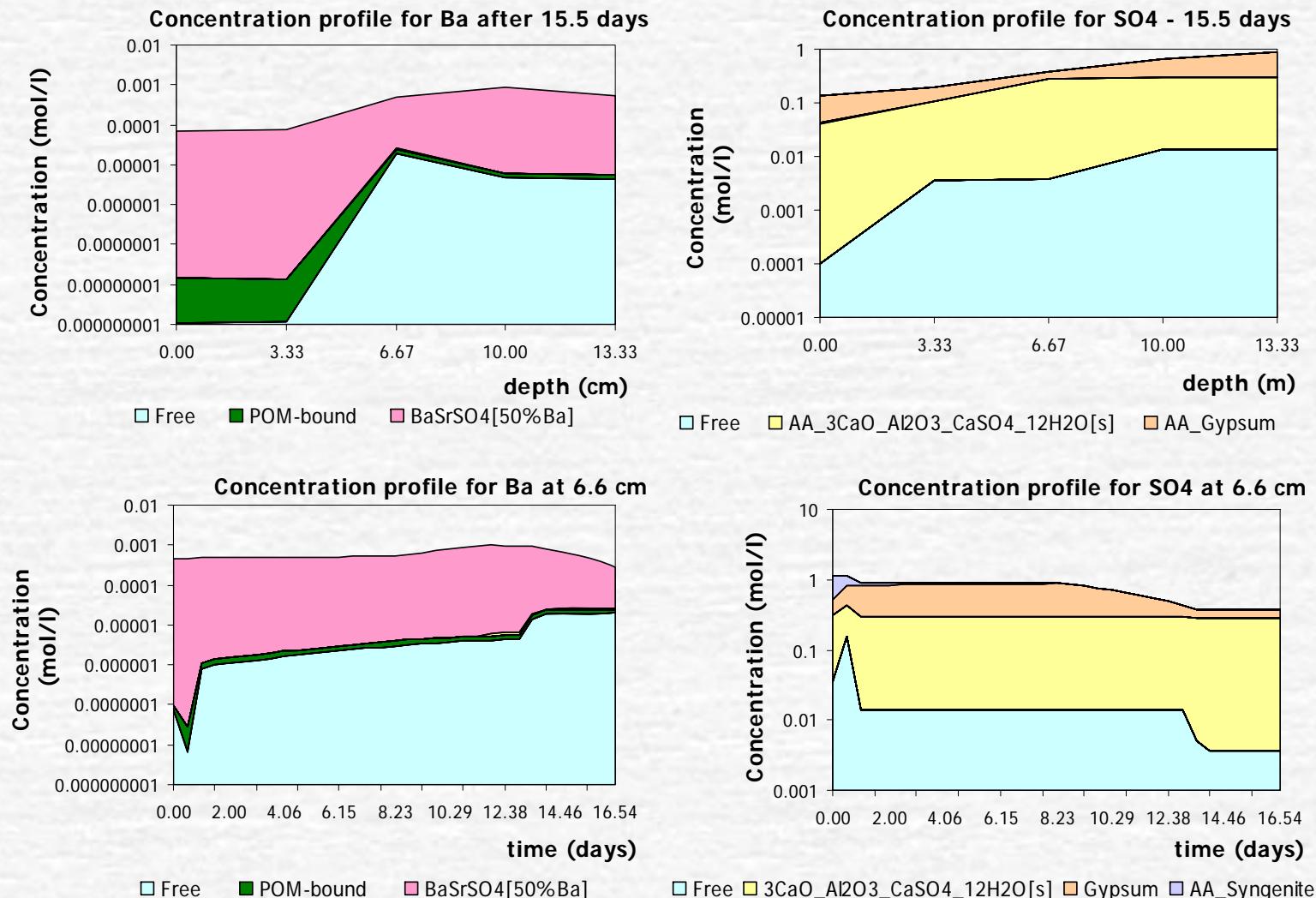


FULL MECHANISTIC MULTIELEMENT PREDICTIVE MODELLING OF PERCOLATION TEST DATA (Cumulative release)



FULL MECHANISTIC MULTIELEMENT PREDICTIVE MODELLING OF PERCOLATION TEST DATA (Partitioning within the column)

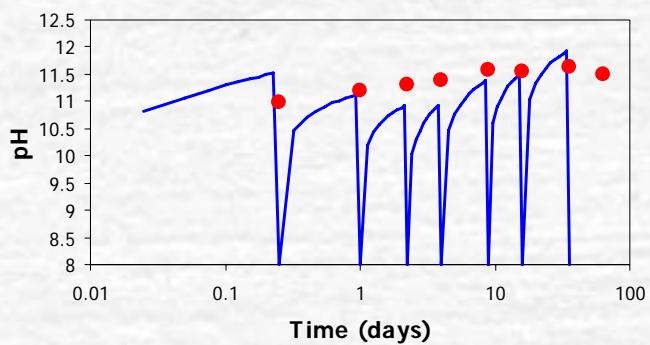
Partitioning in the column as a function of depth at specified time



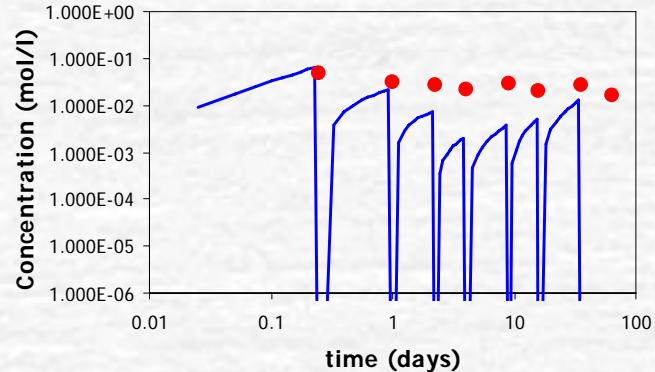
Partitioning in the column as a function of time at specified depth

FULL MECHANISTIC MULTIELEMENT PREDICTIVE MODELLING OF TANK TEST DATA OF A STABILISED WASTE FORM

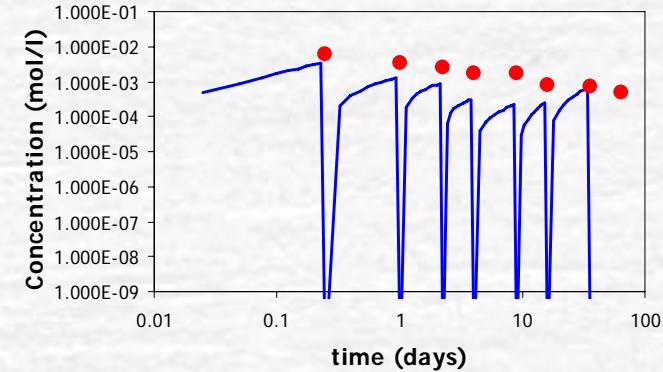
Stabilised waste Tank test:
pH as function of Time



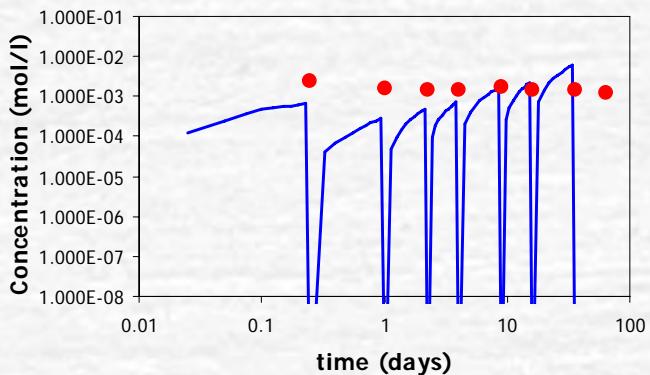
[Na⁺] as function of time



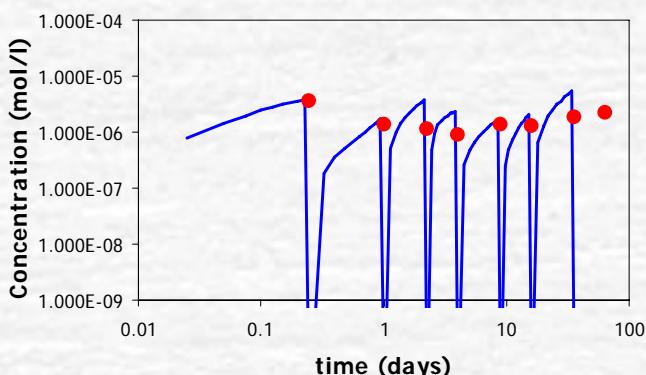
[SO₄-2] as function of time



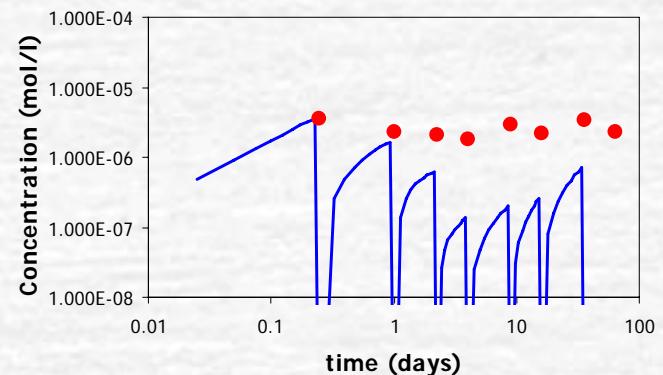
[Ca⁺²] as function of time



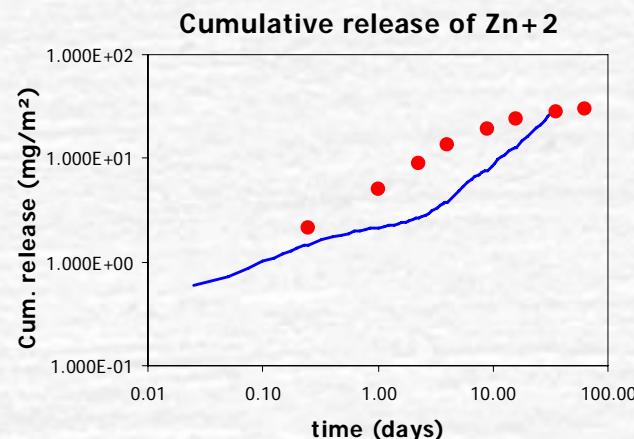
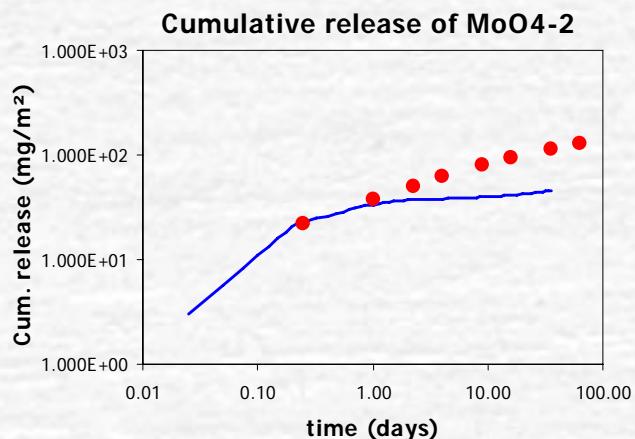
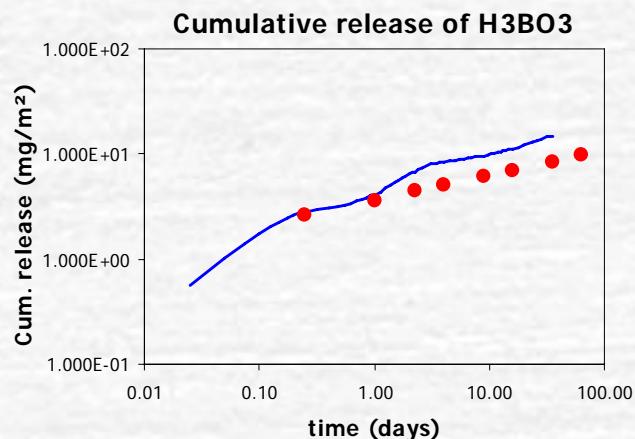
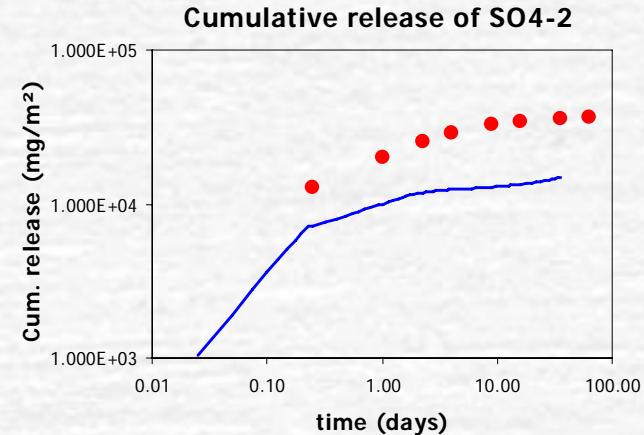
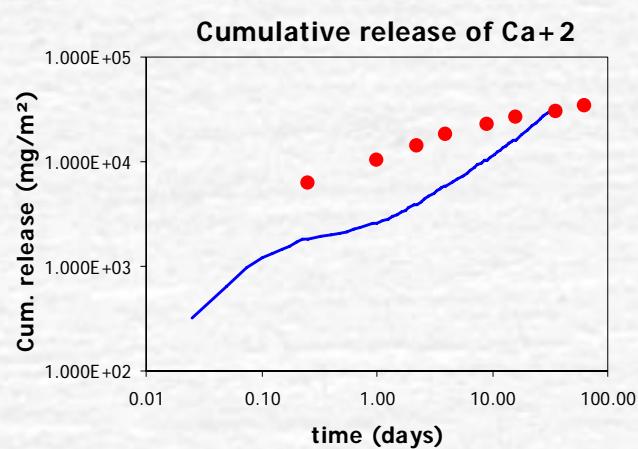
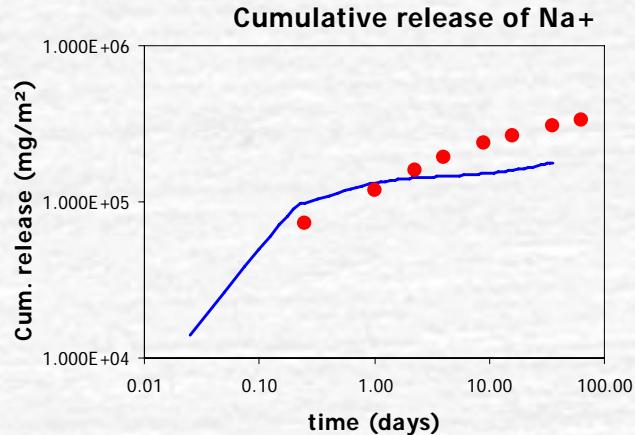
[H₃BO₃] as function of time



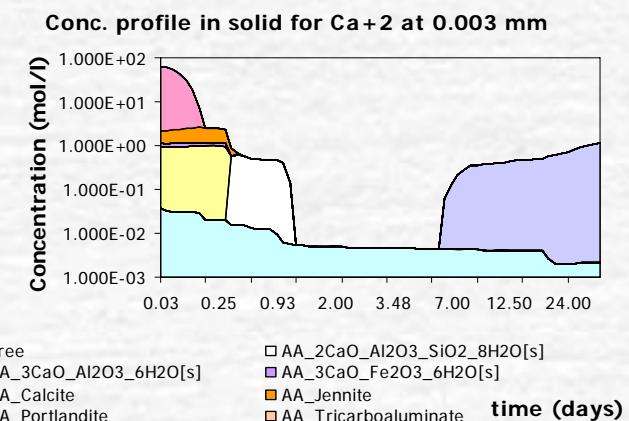
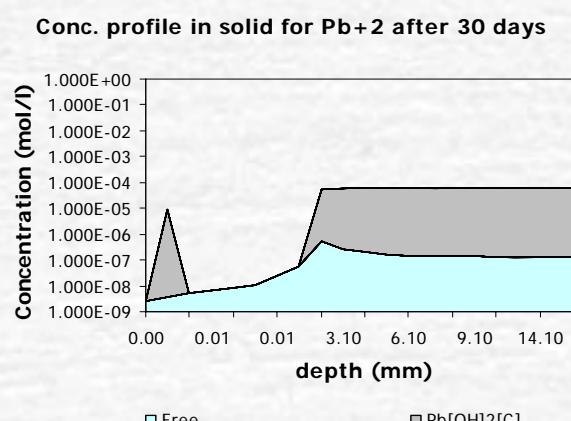
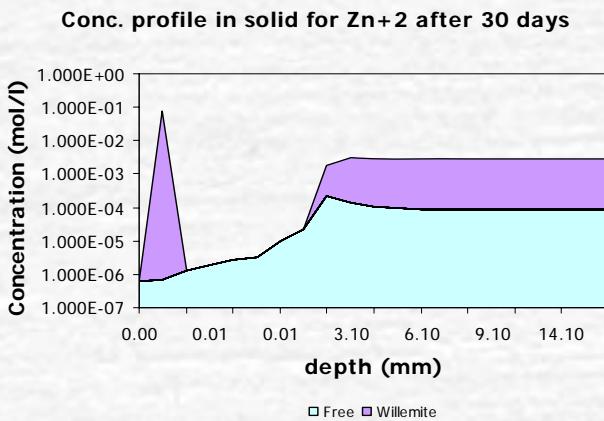
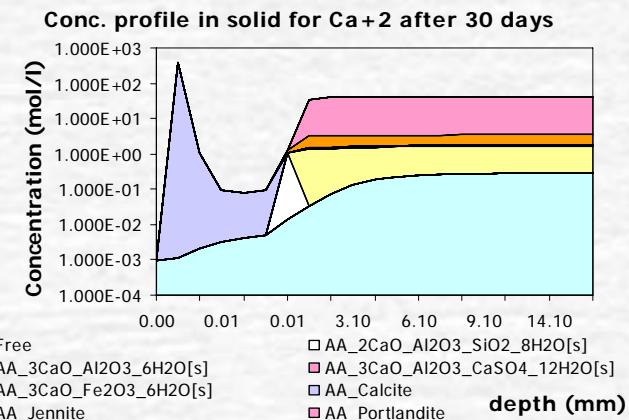
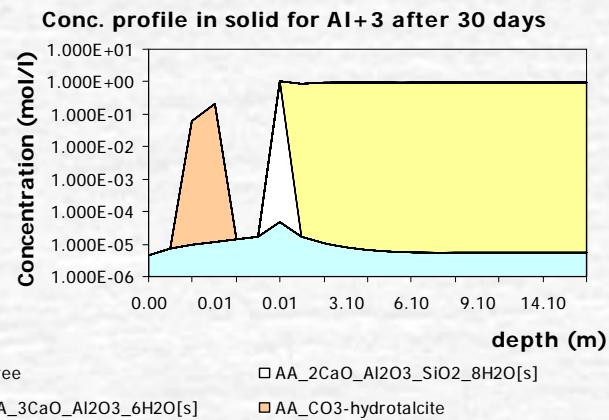
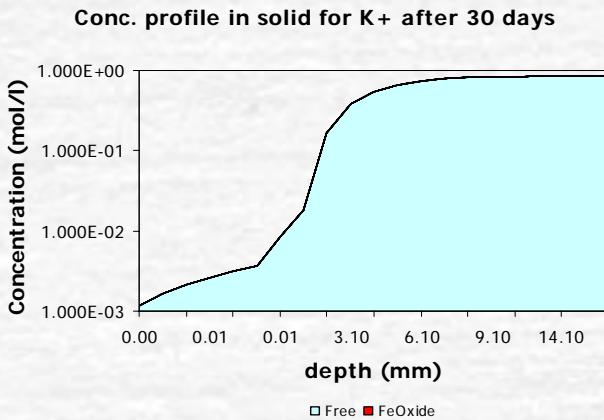
[MoO₄-2] as function of time



FULL MECHANISTIC MULTIELEMENT PREDICTIVE MODELLING OF TANK TEST DATA OF A STABILISED WASTE FORM

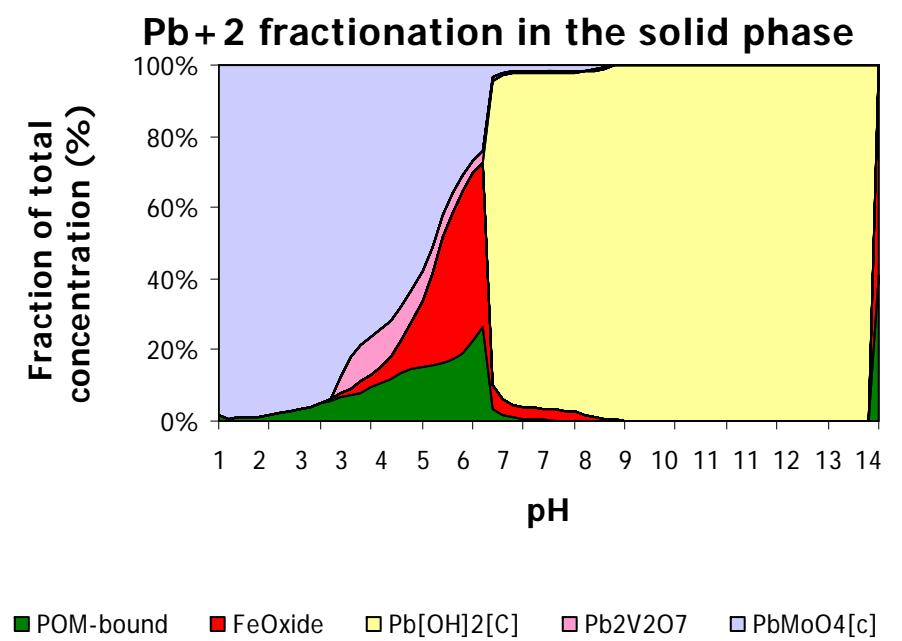
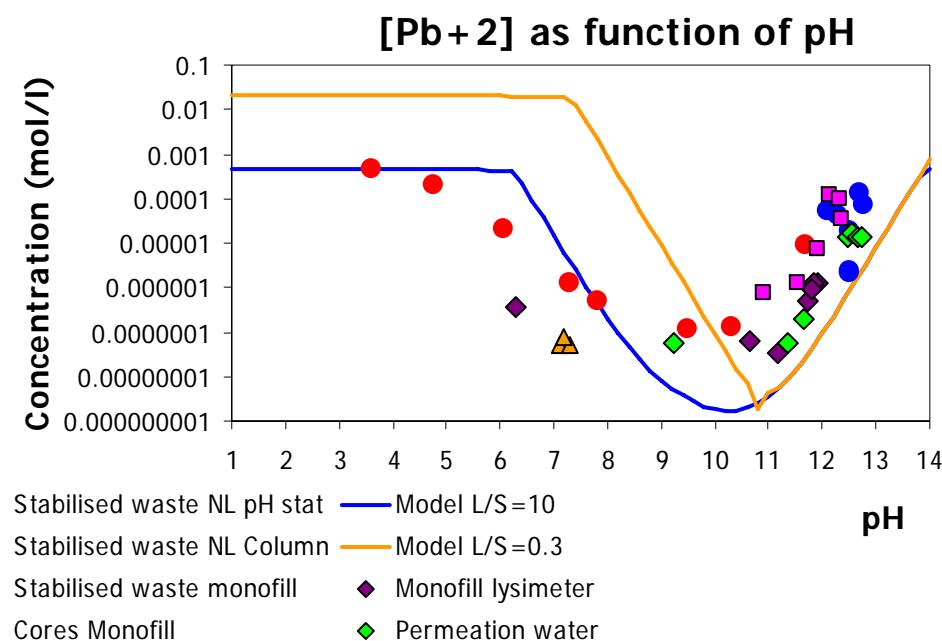


Predicted profile of cement mortar after carbonation



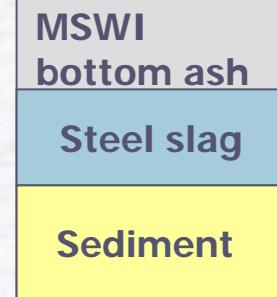
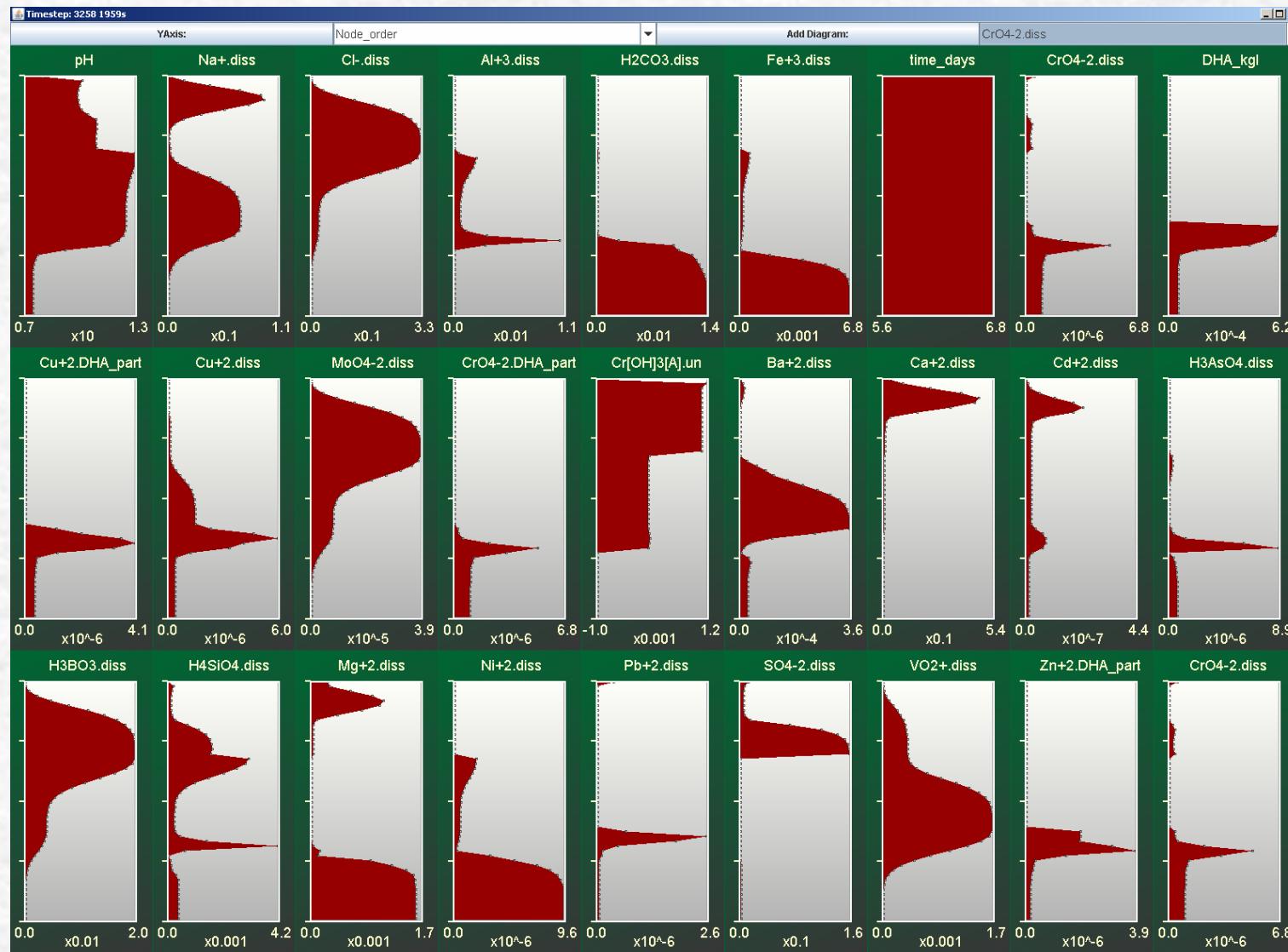
Carbonation affects a thin surface layer of cement mortars

Integration of test results from lab, lysimeter, core sample leaching, field percolate and modelling at a full scale stabilisation plant

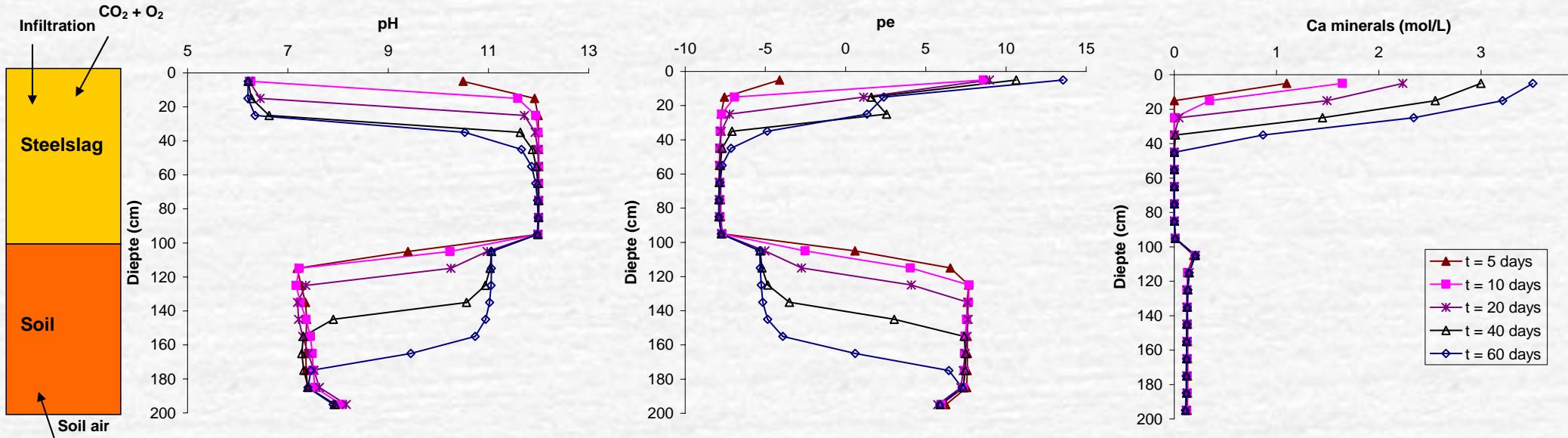


Different testing conditions: Same geochemical controlling processes

Modelled release profiles in a three layer system



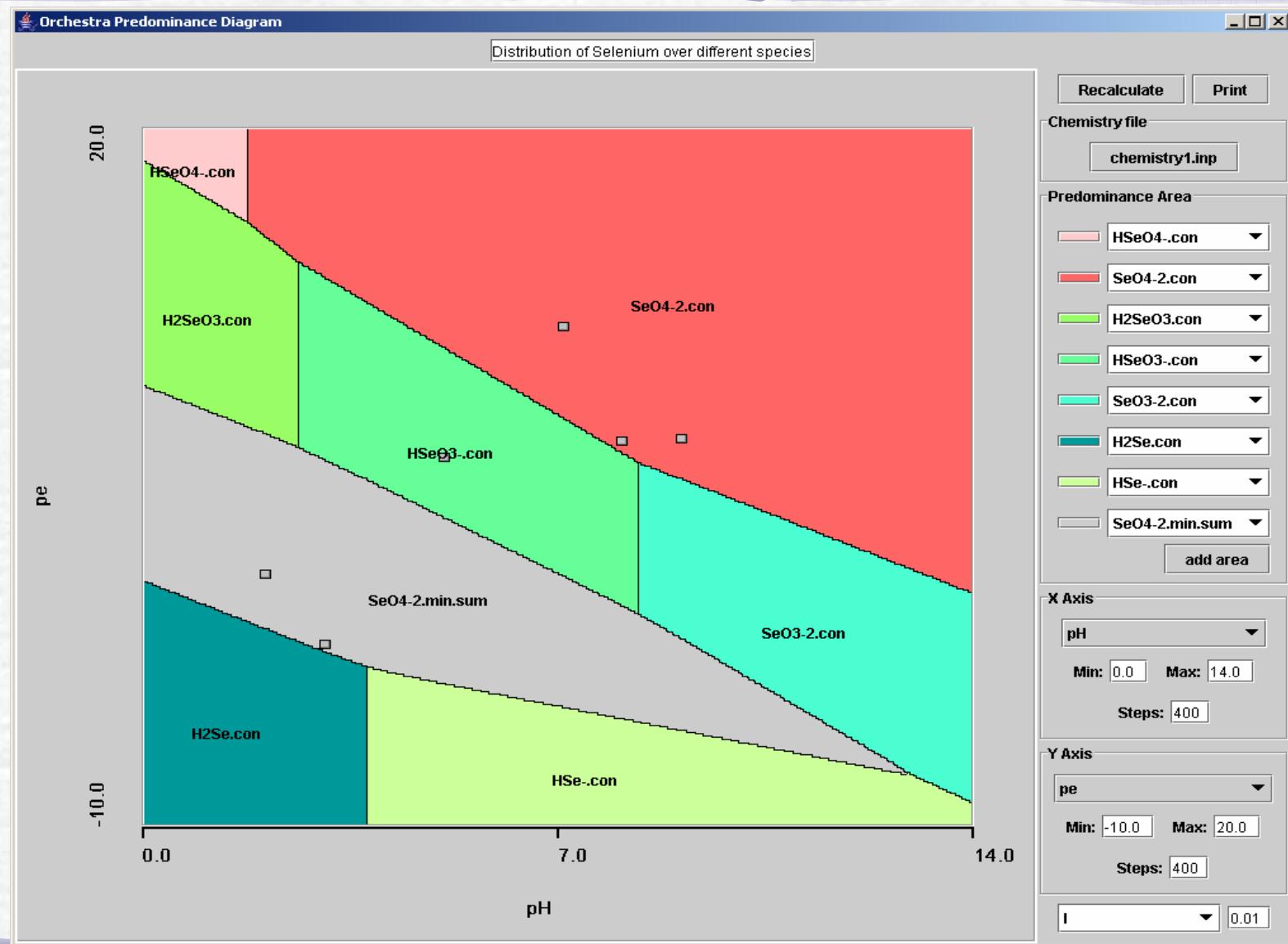
Modeling pH and redox effects from steelslag exposed to the atmosphere



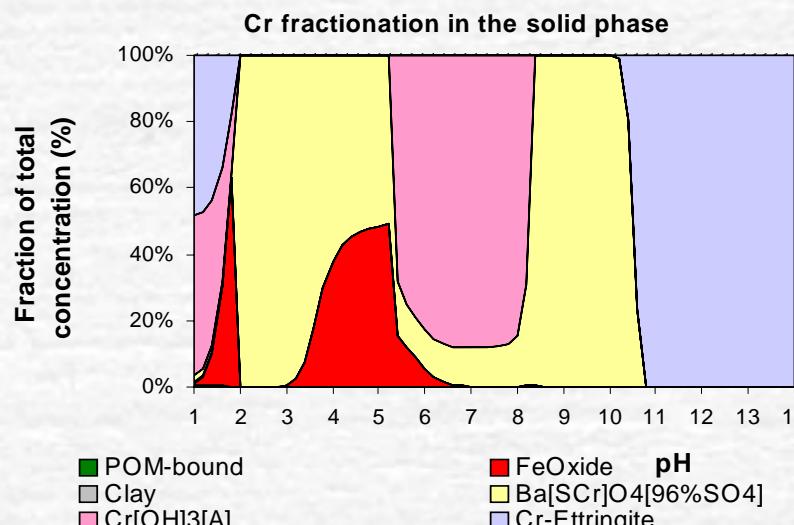
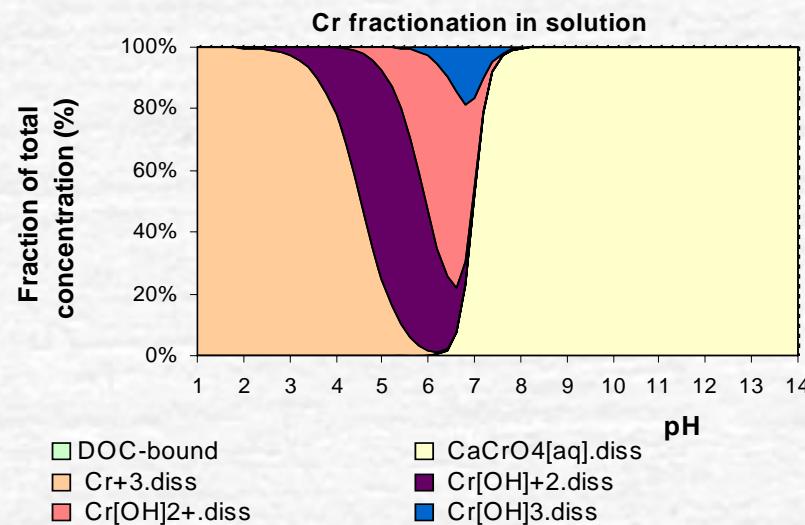
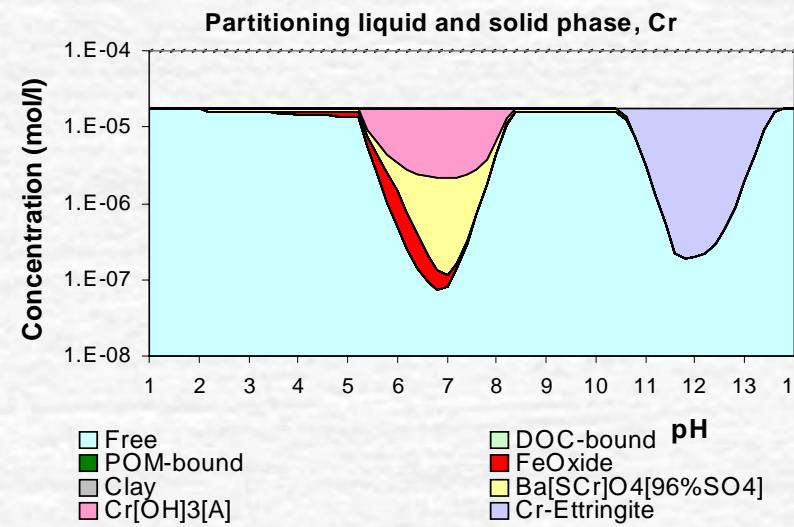
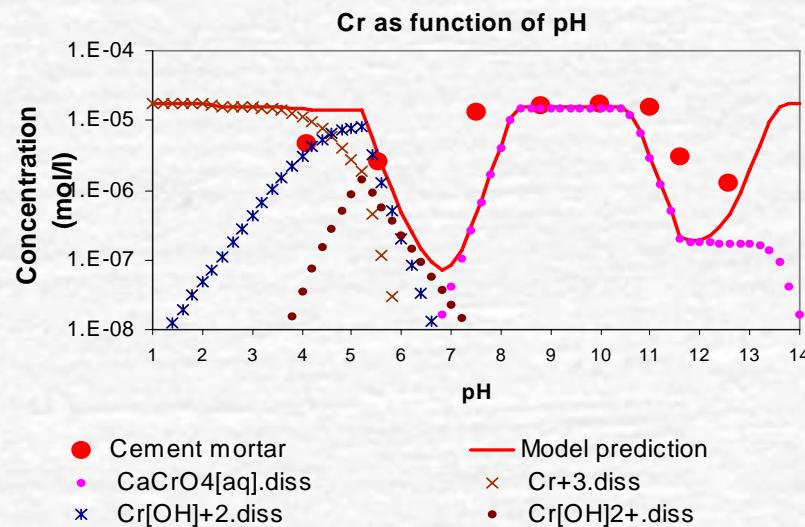
Steel slag : high pH en reducing (sulfides)

Application: Sand like fraction applied in parking lots and industrial parks leading to unacceptable impact on ground and surface water

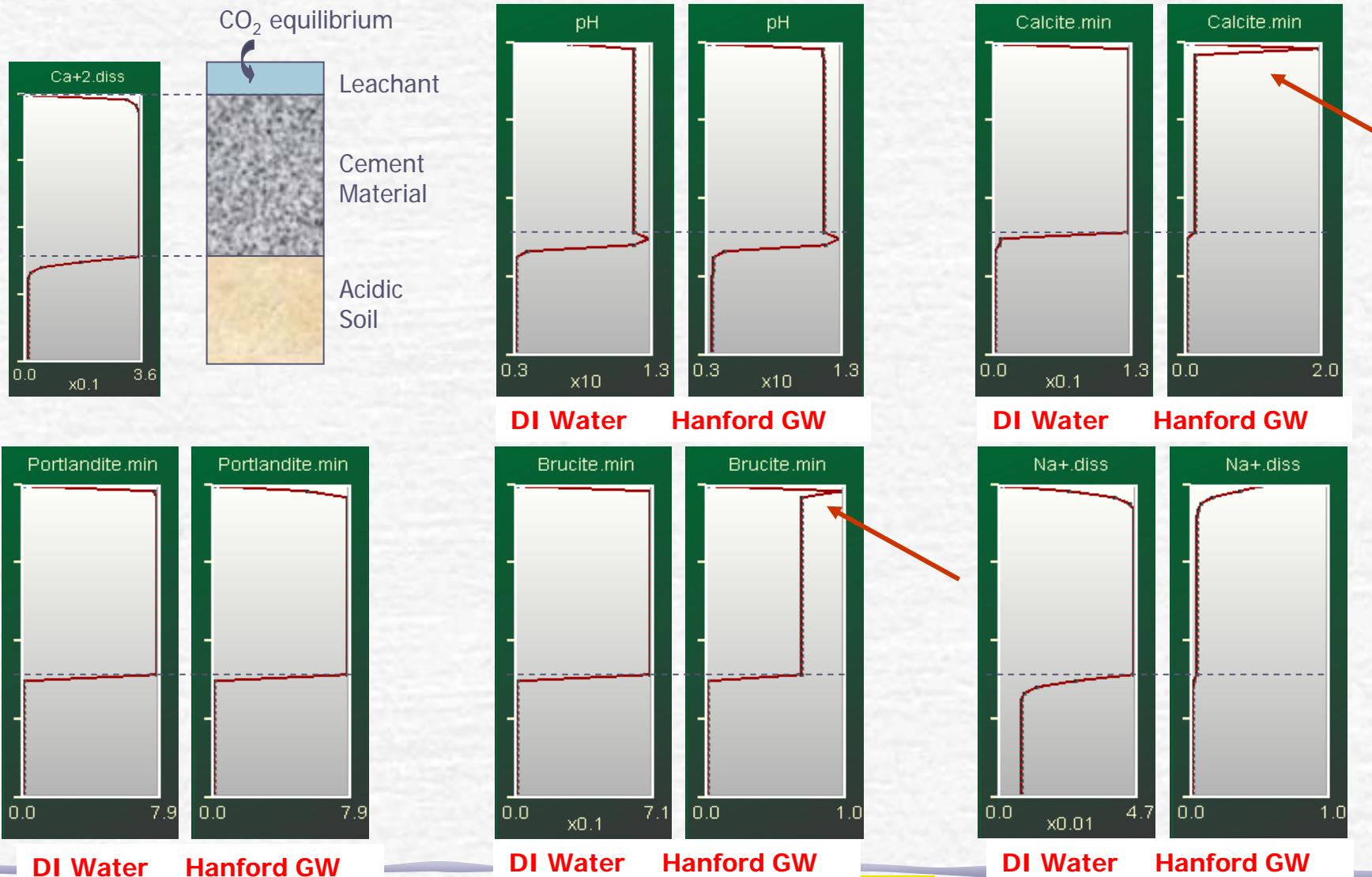
Predominance diagrams in ORCHESTRA



CHROMIUM SPECIATION IN MORTAR AND WATER



Leachant Simulation – Boundary Effects



Scenario Database (in preparation)

- ✓ road construction sub-base and embankments
- ✓ landfill
- ✓ monolithic waste landfill
- ✓ drinking water pipes
- ✓ treated wood construction
- ✓ multilayer applications
- ✓ oxidation of mining waste and slag
- ✓ monolithic structures
- ✓ prediction of release from material mixtures (blended cements, soil-fertiliser, soil-soil improver and mixed wastes)

Conclusions

- The characterisation leaching test methods developed in CEN/TC 292 for waste and ISO/TC 190 Soil are equally applicable to a wide range of other materials such as construction products and stabilised radioactive waste (horizontal standardisation). Similar methods will likely be adopted in SW 846 (US EPA)
- An integrated approach of assessing environmental impact for broad range of materials and products is the way to proceed and will help to avoid unnecessary duplication of work.
- Characterisation as described here will provide better understanding of material behaviour and partitioning and thus provide means to find solutions to
 - improvement of environmental quality,
 - a proper source term for impact evaluation in different exposure scenarios,
 - reference behaviour for typical materials
 - bioavailability and
 - allow placement of other (more limited) test data in perspective.
 - prediction of material mixtures

Conclusions

- The chemical speciation fingerprint derived from the pH dependence test provides a chemical speciation fingerprint suitable for prediction of different laboratory test results, more complex impact modeling, evaluation of material mixes and a basis for assessing multilayer scenarios.
- The insight in release controlling processes provides better means to derive optimal compliance test procedures. Placing compliance test data in conjunction with characterisation data allows more far reaching conclusions.
- The full mechanistic modelling approach presented here is an ambitious approach, but given the current software capabilities it definitely seems the best way forward to environmental impact modelling of the wide variety of materials potentially affecting the atmosphere, soil, ground and surfacewater.
- Further development will be focused on building new scenarios and the verification of field observations against model predictions.

ACKNOWLEDGEMENT

This work has been supported partly by DOE/CRESP
and by A&G Maasvlakte (formerly VBM),
Rozenburg, The Netherlands



INFORMATION ON LEACHING AVAILABLE AT:

LEACHING BACKGROUND

www.leaching.net (Wascon 2003 workshop Leaching)

CONSTRUCTION PRODUCTS DIRECTIVE

www.cenorm.be/cenorm/workarea/sectorfora/construction+sector+network/conference.asp (CEN Workshop)

Report ECN-C--05-45 and ECN-C--04-060

LEACHING IN PROJECT HORIZONTAL

www.ecn.nl/horizontal (Desk study 23)

GRACOS EU project on contaminated soil and sediment

www.uni-tuebingen.de/gracos (Guideline)

Additional Background Slides

pH DEPENDENCE TEST

pH DEPENDENCE TEST TO ASSESS SENSITIVITY
TO CHANGES IN pH - pH stat and "ANC" mode

TEST CONDITIONS **ANC MODE:**

BATCH TEST

8 FINAL pH VALUES (pH 4-12)

LEACHANT: PREDETERMINED AMOUNTS OF ACID
OR BASE

LIQUID TO SOLID RATIO (L/S) = 10

RESULTS IN mg/l (GEOCHEMICAL MODELLING) OR
mg/kg (RELEASE EVALUATION)

Scenario
Description

Material
characterization

Controlling
factors

Modelling
leaching

Validation
verification

Evaluation

Conclusions

pH DEPENDENCE TEST TO ASSESS SENSITIVITY TO CHANGES IN pH, E_H AND TEMPERATURE

(PrEn14429 Batch mode test)



CEN/TC 292 -
EN 12920

Scenario
Description

Material
characterization

Controlling
factors

Modelling
leaching

Validation
verification

Evaluation

Conclusions

ADVANTAGES OF pH DEPENDENCE TEST

- Identification of sensitivity of leaching to small pH changes
- Provides information on pH conditions imposed by external influences
- Basis for comparison of international leaching tests
- Basis for geochemical speciation modelling
- Provides acid neutralization capacity information
- Mutual comparison of widely different materials to assess similarities in leaching behaviour
- Recognition of factors controlling release
- For non-interacting species possible to assess sub-sampling error

Applicable to almost any material

PERCOLATION TEST TO ASSESS LONG TERM RELEASE FOR GRANULAR MATERIALS TS 14405

Liquid to solid ratio (L/S) related to a time scale by infiltration rate, density and height of application.

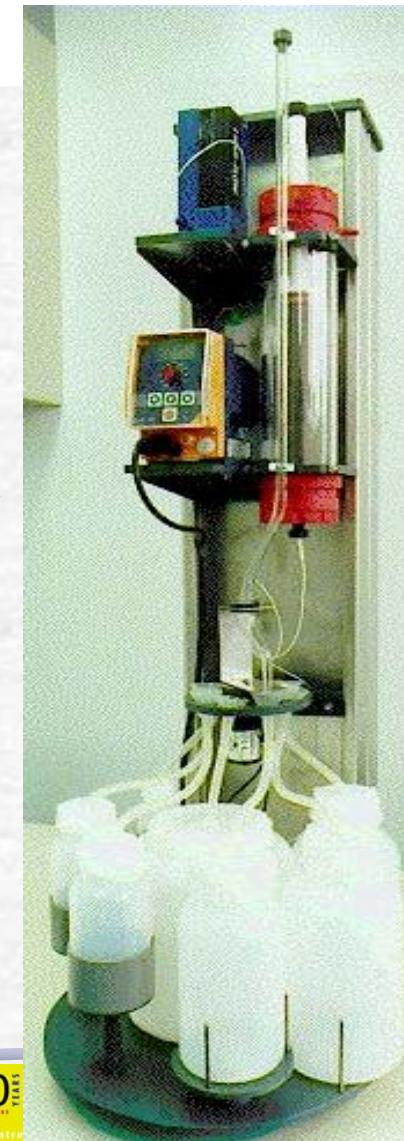
TEST CONDITIONS:

Pre-equilibration after saturation for more than 48 hrs

Up-flow

L/S range 0.1 - 10 (100 - 1000 yrs)

Test data in mg/l or mg/kg cumulative



ADVANTAGES OF PERCOLATION TEST

- Identification of solubility control versus wash out
- Indication of pore water concentrations relevant to field leachate from low L/S data
- Local equilibrium established quite rapidly
- Basis for geochemical speciation modelling
- Allows comparison with lysimeter and field data provided L/S value can be obtained from such measurements
- Projection towards long term behaviour possible

Solubility controlled release

Wash-out of non-interacting species

Applicable to many materials. Limited or not applicable to clayey soils and sediments (low permeability).

TANK LEACH TEST OR COMPACTED GRANULAR LEACH TEST (CGLT) FOR MONOLITHIC MATERIALS (modified)

TEST CONDITIONS:

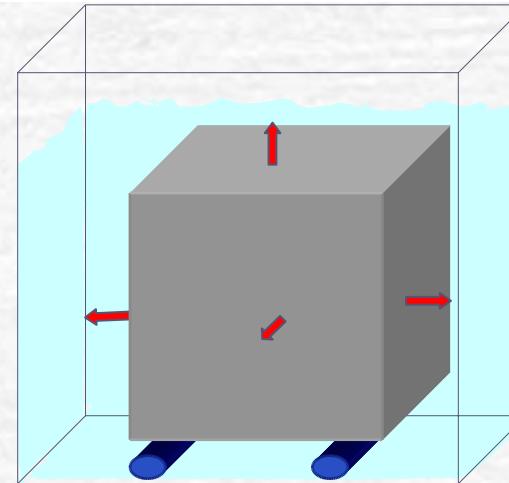
First step: pre-equilibration
for 48 hrs at liquid to
volume ratio: 5

Second step: leaching
at low L/V ratio (1) for 24 hrs

Then contact times: 2, 4, 8, 16, 32 and 64 days

Leachant: demineralised water (own pH)

Expression of results in mg/m² (cumulative)
against time



CEN/TC 292 -
EN 12920

Scenario
Description

Material
characterization

Controlling
factors

Modelling
leaching

Validation
verification

Evaluation

Conclusions

EXPERIMENTAL SET-UP



CGLT = Compacted Granular Leach Test

ADVANTAGES OF TANK LEACHING TEST OR COMPACTED GRANULAR LEACH TEST

- Relevant for materials with monolithic character (durable materials) or materials behaving as monolith (low permeability soil and sediments)
- Identification of solubility control versus dynamic leaching possible
- Isolation of surface wash-off effects
- Quantification of intrinsic release parameters
- Basis for reactive/transport modelling
- Projection towards long term behaviour possible

Applicable to sediments, clayey soils, stabilised materials and construction materials produced